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Thermal conductivity of liquid metals.

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Brown University

THERMAL CONDUCTIVITY OF LIQUID METALS

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Abstract of

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by John Ives Kineke, M.Sc., Brown University, June 1967

An apparatus is designed and built to measure the thermal conductivity of various liquid metals at temperatures up to 400°C. The method of longitudinal heat flow with guard ring compensation is used. Measurements on liquid tin at 298°C indicate its thermal conductivity to be 0.0802 cal/sec/cm/cm^2/°C (cgs units) which agrees with previous experiments. The experimental error in the present work is just over 2%. A review of significant previous work is also given.

THERMAL CONDUCTIVITY OF LIQUID METALS

by

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B.E.E., University of Louisville, 1958

Thesis

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I. INTRODUCTION

An investigation of the currently available literature concerning the measurement of thermal conductivity of metals in the liquid state yields a relatively small amount of information. Measurements of this type have been made by various investigators dating back to 1917, but a comparison of the several data does not show good agreement in many cases. The situation was appropriately stated by Cusak[1] in 1962: "---the literature not infrequently stresses the importance of thermal conductivity and goes on to deplore the scarcity of reliable values. This still seems to be the position."

The purpose of the present work was to design and build an apparatus for experimental determination of the thermal conductivity of any liquid metal at temperatures not exceeding 400°C. As will be seen later, the design is closely modeled after that used by Ewing[2] because of the high accuracy of his results. The device has been used to measure the thermal conductivity of liquid tin at several temperatures, and the results show fairly good agreement with some previous experiments.

Thermal conductivity measurements have an important application in the study of acoustical absorption in liquid metals. The classical Stokes-Kirchhoff theory of acoustical absorption indicates that the absorption of sound is due to losses of energy conducted away in the form of heat and the frictional losses of viscous flow. More recent studies have shown that a relaxation process also contributes to this energy loss. In order to determine the energy loss due to the relaxation, the classical absorption is subtracted from the experimentally measured values. In order to determine

the classical value accurately, precise values of thermal conductivity must be known for the metal or alloy under study.

II. BACKGROUND

Experiments to measure thermal conductivity fall into two general categories: The Forbes Bar method and the method of longitudinal heat flow with guard tube compensation. In the former, a thermal gradient is set up vertically in a well insulated liquid sample by means of a heater at the upper end of the sample. The total radial heat loss by the sample is then calculated, assuming the thermal conductivity of the insulation to be known. The sample heater input power is then corrected by this amount. Temperature gradients in the sample are usually measured by suitably calibrated thermocouples; the cross-sectional area of the sample is, of course, known. The thermal conductivity is then calculated using the expression:

$$Q = KA \frac{dT}{dx} \qquad (1)$$

where Q is the heat input to the sample in calories per second, A the area of the sample in square centimeters, $\frac{dT}{dx}$ the thermal gradient in degrees centigrade per centimeter and K the thermal conductivity in calories per second per centimeter per square centimeter per ^{o}C (hereafter called cgs units). In the longitudinal heat flow method, a constant heat flow is set up in a vertical column containing the sample by a heater at the top of the column. The column is surrounded by a metal cylinder or guard tube in which a similiar heat flow is established. If the temperatures and the temperature gradient in the guard tube are equal to that in the sample column, then there is no radial

heat loss. The heat input to the sample column is measured and the gradient again determined by thermocouples. Thus the area is known, the thermal conductivity can be computed by using Eq. (1). Note that in both methods heat is introduced into the liquid sample at the top and flows from top to bottom. This arrangement is necessary to prevent heat transfer through the liquid by convection. For good results, it is necessary that a steady-state condition of heat flow be established in both methods, which usually requires a great deal of time for the measurements.

The first significant work on thermal conductivity of liquid metals was contributed in 1917 by Northrup and Pratt.[3] Their experiment, although actually designed to test the Wiedemann-Franz ratio through a change of state, did furnish values for thermal conductivity of liquid tin* and bismuth. The method consisted of comparing the temperature gradient between two points in a column of the metal being tested with the gradient in a coaxially mounted steel cylinder. Heat is uniformly supplied to the sample by an electrically heated copper bar and the opposite end of the steel rod is immersed in boiling water. The temperature gradients are measured by thermocouples imbedded in both sample and steel. The results obtained from this device are a ratio of the thermal conductivity of the liquid metal sample to that of the steel. In the measurements it is assumed that the radial heat loss is the same from the sample and steel. The results for tin shown on Fig. 12 are computed by assuming a value of thermal conductivity of the steel.

In 1920, Konno[4] made a large number of thermal conductivity measurements on tin, lead, bismuth, zinc, aluminum and antimony in the solid and liquid states. Konno's apparatus was similar to that of Northrup, using cylinders

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* The results of the experiments discussed are all shown in Fig. 12 for liquid tin.

of steel mounted coaxially above and below the liquid sample. The radial heat loss was calculated and temperature gradients measured by iron-nickel and platinum-rhodium thermocouples. Konno's measurements were made at temperatures up to 800°C which are the highest found. The results of this comprehensive study showed that for all metals tested the thermal conductivity drops abruptly on melting, and then decreases slightly with increasing temperature.

In 1923, Brown[5] measured the thermal conductivity of solid and liquid tin, cadmium, thallium and tin, lead, zinc and bismuth alloys. His apparatus was of the longitudinal heat flow type and consisted of a slate tube of the material under test mounted vertically in the center of a hollow cylinder or guard ring of brass. The tops of the sample column and outer cylinder were heated by electric heating coils, and similar thermal gradients are set up in each. Since the temperature at any given height in the apparatus is constant, radial heat loss from the sample column is eliminated, and the conductivity can be computed from the standard conduction equation if the heat input, area and temperature gradient of the sample are measured. In Brown's experiment, since a slate tube was used as the holder of the liquid metal sample, the quantity of heat conducted through the slate was calculated and subtracted from the total heat supplied by the specimen heater. Brown's results, shown on Fig. 12, agree fairly well with Konno's, but are somewhat higher than those shown for Northrup and Pratt. This is attributed to the fact that Northrup's apparatus provided no compensation for radial heat loss resulting in an inherent error in his measurements.

In 1938, Hall[6] measured thermal conductivities of liquid mercury, sodium and sodium amalgams. His apparatus, like Brown's, had the liquid metal sample contained in a central vertical column surrounded by two metal cylinders,

which acted as a guard ring. The temperature of these cylinders was measured by two resistance thermometers, and the ratio of their resistances was kept constant and equal to its equilibrium value at room temperature. This arrangement prevented radial heat loss from the liquid sample. A rather clever automatic device was used to keep the temperatures of the guard cylinders constant. Hall's measurements were all made at temperatures less than 220°C and his apparatus was not adaptable for use above this temperature.

Bidwell[7] devised an apparatus of the Forbes bar type for thermal conductivity measurements at temperatures up to 730°C. The sample was held in a hollow graphite rod mounted vertically inside an iron guard cylinder, with silocel packed in between. After the entire apparatus was raised to sufficiently high temperature a heater at the top of the liquid sample was energized and a gradient established in the liquid metal. The guard ring was not heated in such a way that radial heat flow was eliminated, and consequently it was necessary to compute the radial heat loss, which could be done if the thermal conductivity of the silocel insulation was known. The results of this experiment compared quite well with Konno's for zinc. Bidwell felt very strongly that his apparatus would give the most accurate thermal conductivity measurements and stated "---the author ventures the opinion that (his method) is so far superior to any hitherto reported --- that it should become standard." This was shown to be a minority opinion by other workers. Later, in 1940, Bidwell[9] published another paper describing measurements in liquid and solid lead using the above apparatus modified to include an additional iron cylinder which served as a second guard ring and reduced radial heat flow almost to zero. In the same paper, Bidwell outlined an "intercept theory" which allowed the thermal conductivity of metals in the liquid state to be estimated. Using

a classical argument, he derived the relation

$$\frac{K}{\rho c} = \frac{k}{T} + k'$$

where K is the thermal conductivity, ρ the density, c the atomic heat, T the temperature and k, k' are constants. Bidwell showed that if one plotted $\frac{K}{\rho c}$ vs. $\frac{1}{T}$ for a solid metal, the intercept at $\frac{1}{T} = 0$ would be the proper value of $\frac{K}{\rho c}$ for the liquid state. He then showed that Konno's thermal conductivity results for tin and his own values for lead and zinc verified the theory. In 1947, Powell[9] pointed out some rather glaring inconsistencies in Bidwell's observations and in a later article[10] suggested that the data used to verify the relation may have been selected. Although it appears that Bidwell's intercept theory is not a good one for estimating thermal conductivities of liquid metals, it deserves credit as the only one ever proposed on this subject. In addition to disposing of Bidwell's intercept theory, Powell published in 1949[10] a summary of all thermal conductivity data for liquid metals available at that time.

The U.S. Naval Research Laboratory sponsored work in 1952 by Ewing,[2] Grand and Miller on the measurement of thermal conductivity of liquid sodium, potassium and some of their alloys. This group selected the longitudinal heat flow method for its greater accuracy and designed an apparatus which gave results with errors less than 2%. It is this apparatus which was used as a model for the equipment built in the present work. Ewing's apparatus was later[11] (1954) used for measurements in mercury and two additional sodium-potassium alloys, again with good precision.

The most recent thermal conductivity measurements were made by Pashaev,

who studied the changes in thermal conductivity of metals on melting.[12,13] The materials studied were tin, bismuth, gallium and alloys thereof. The complete description of Pashaev's apparatus is in another paper[14] which has not been translated from Russian, but from his comments it appears that the longitudinal heat-flow method is used. Pashaev claims to achieve accuracies to within 5% in his measurements.

III. CONSTRUCTION OF THE APPARATUS

A. General

As previously mentioned, the thermal conductivity apparatus used in this experiment was patterned after that of Ewing, Grand and Miller. A cross sectional view of this equipment is shown in Fig. 1. The sample column or specimen bar (1), guard ring (2), melting chamber (3) and top and bottom covers (4) are all fabricated from type 304 stainless steel. This material was chosen because of its high resistance to corrosion and good machining qualities. This entire assembly is surrounded by the furnace core (16), which is insulated with about three inches of granular alundum and has outer walls of 1-inch Marinite* (not shown on the diagram). The entire apparatus weighs about 100 pounds.

B. Specimen Bar Assembly

The specimen bar assembly, comprised of four separate sections, is 1 5/8 inches in diameter and 20 inches long when assembled. The two lower parts which are the specimen bar proper, (1a & 1b) bolt together with eight 8-32 x $\frac{1}{2}$ allen head cap screws. The joint between sections is gasketed with a 1/32-inch Raybestos-Manhattan type A-36 pressed asbestos flat ring gasket,

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* Trade mark - Johns-Manville Co.

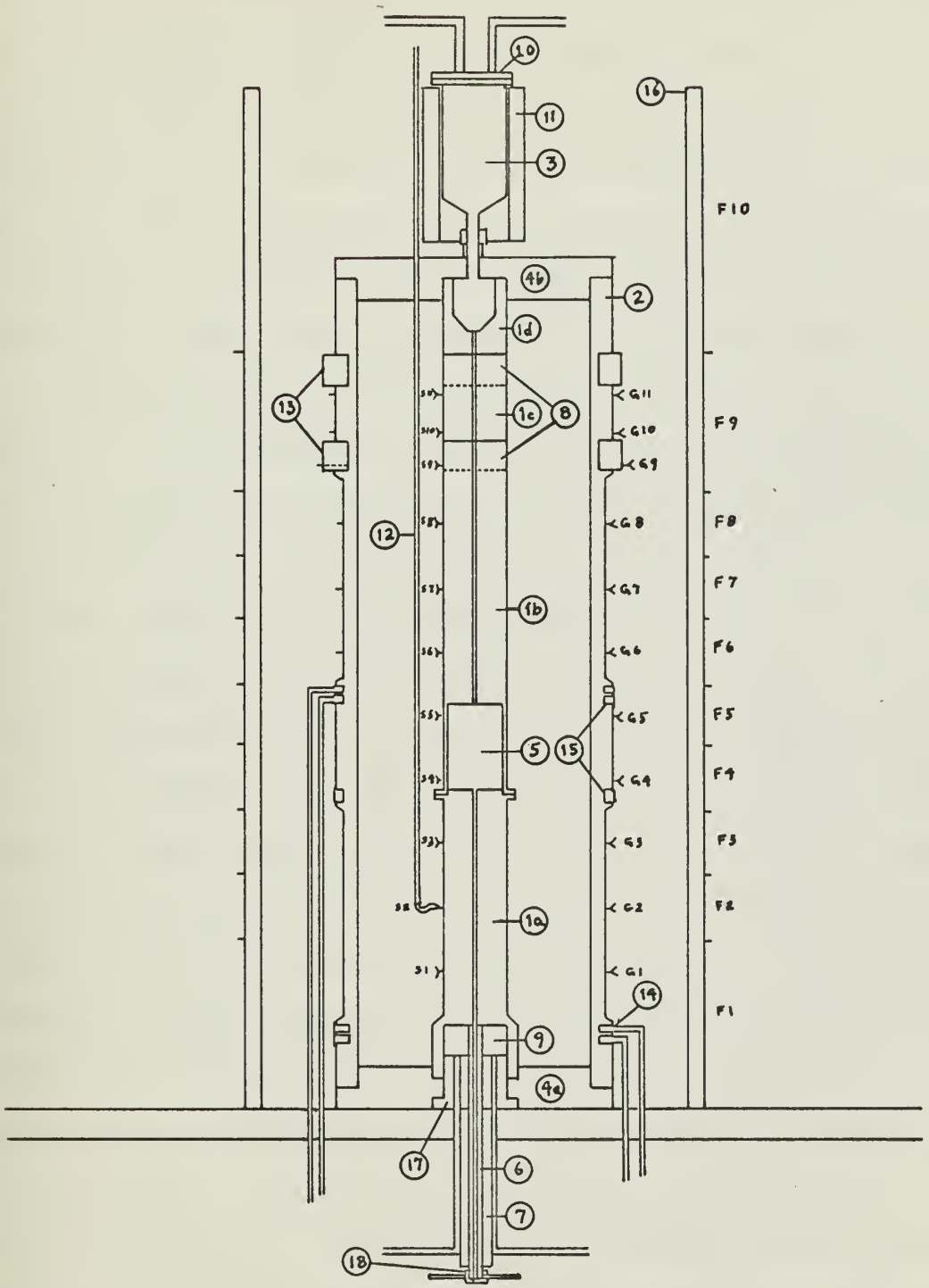

Fig. 1. Cross sectional view of thermal conductivity apparatus. 1a-1d. Specimen bar; 2. Guard Ring; 3. Melting chamber; 4a, 4b. Bottom and top covers; 5. Specimen chamber; 6. Drain tube 7. Drain tube heater; 8. Specimen Bar heaters; 9. Cooling cavity; 10. Melting chamber lid; 11. Melting chamber heater; 12. Thermocouples protection tube (only 1 of 6 shown); 13. Guard ring heaters; 14. Guard ring cooling grooves; 15. Supplementary guard ring heating and cooling; 16. Furnace core; 17. Base plate plug 18. Drain tube cap; S1-S11 Specimen bar thermocouples; G1-G11 Guard ring thermocouples; F1-F10 Furnace core heater segments.

and sealed with Never-Seez* compound. The asbestos gasket and its sealant are capable of withstanding temperatures of 600° C. The wall thickness of the specimen chamber (5) is machined to 1/32 inch which reduces the amount of heat not conducted through the sample to a minimum. A 1/8-inch diameter hole is drilled from top to bottom of the specimen bar to allow for filling and draining of the liquid metal samples. A 1/4-inch stainless steel drain tube (6) is welded to the lower section and passes out through the bottom of the apparatus. It is surrounded by a heater (7) to keep the tube above the melting point of the liquid metal sample when draining and filling. The bottom surface of the specimen chamber is machined slightly concave, so that the liquid metal will drain out completely. Heat is supplied to the specimen bar by two specially built electric heaters mounted near the top of the bar (8). The donut shaped heaters fit in a space 1 1/2 inches in diameter and 7/8 inch high. A disc of pure copper is silver-soldered tightly into the bottom of each heater space, providing even distribution of heat over the cross-section of the specimen bar. As shown in Fig. 1, one heater is located at the top of the upper section (1b) of the specimen bar proper, and the other is 1 1/2 inches above the first, mounted in a 1 5/8-inch diameter spacer (1c). Heat is removed from the specimen bar by the introduction of cooling air to the space hollowed out for that purpose near the bottom (9). The topmost part of the specimen bar, called the expansion chamber (1d), is hollowed out to allow for expansion of the liquid metal as it is heated. The expansion chamber fits into the top cover (4b) and is held in place by six 10-32 x 1/2 allen head cap screws. This joint is also gasketed with 1/32 inch pressed asbestos and sealed with Never-Seez. The melting chamber (3) is fastened

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* Trade mark - Never-Seez Corp.

to the top cover by a 3/8-inch Gyrolok* connector. The lower part of the Gyrolok connector is heli-arc welded to the top cover to assure a good seal. The melting chamber lid (10) has two 1/4-inch Gyrolok connectors welded to its upper surface, and these provide seals for the 1/4-inch tubing leading to the vacuum system. The melting chamber is surrounded by another specially made electric heater (11) which provides heat to melt the specimen. The specimen bar assembly is pictured completely assembled in Fig. 2.

The entire inside of the specimen bar assembly, including the melting chamber, is sealed and is capable of holding a vacuum pressure of 50 microns. This capability is accomplished by the compressed asbestos gaskets previously described and by a rather unique copper ring seal** located just below the expansion chamber. Figure 3 is an exploded cross-sectional view of this sealing arrangement, and the upper specimen bar assembly. A 1/4-inch O.D. stainless steel tube is butt-welded into the upper section of the specimen bar proper. This tube extends through the heater spacer and threads into the expansion chamber. The copper ring seal fits snugly between the expansion chamber and heater spacer. When the expansion chamber is tightly screwed down, the ring seal is flattened downward and inward against the steel tube and a tight vacuum seal is thus accomplished. The specimen bar heaters, of course, fit into the donut shaped spaces in the specimen bar upper section and heater spacer.

The construction of the heaters for the specimen bar, melting chamber and drain tube is somewhat unique and deserves comment. These heaters consist of 1/8-inch diameter coils of No. 22 Kanthal type A-1 resistance wire which are held in place by Sauereisen No. 6 electric heater cement. The specimen bar

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* Trade mark - Hoke Mfg. Co.

** Designed and built by R. Waters and C. Garber of the Brown University instrument shop.

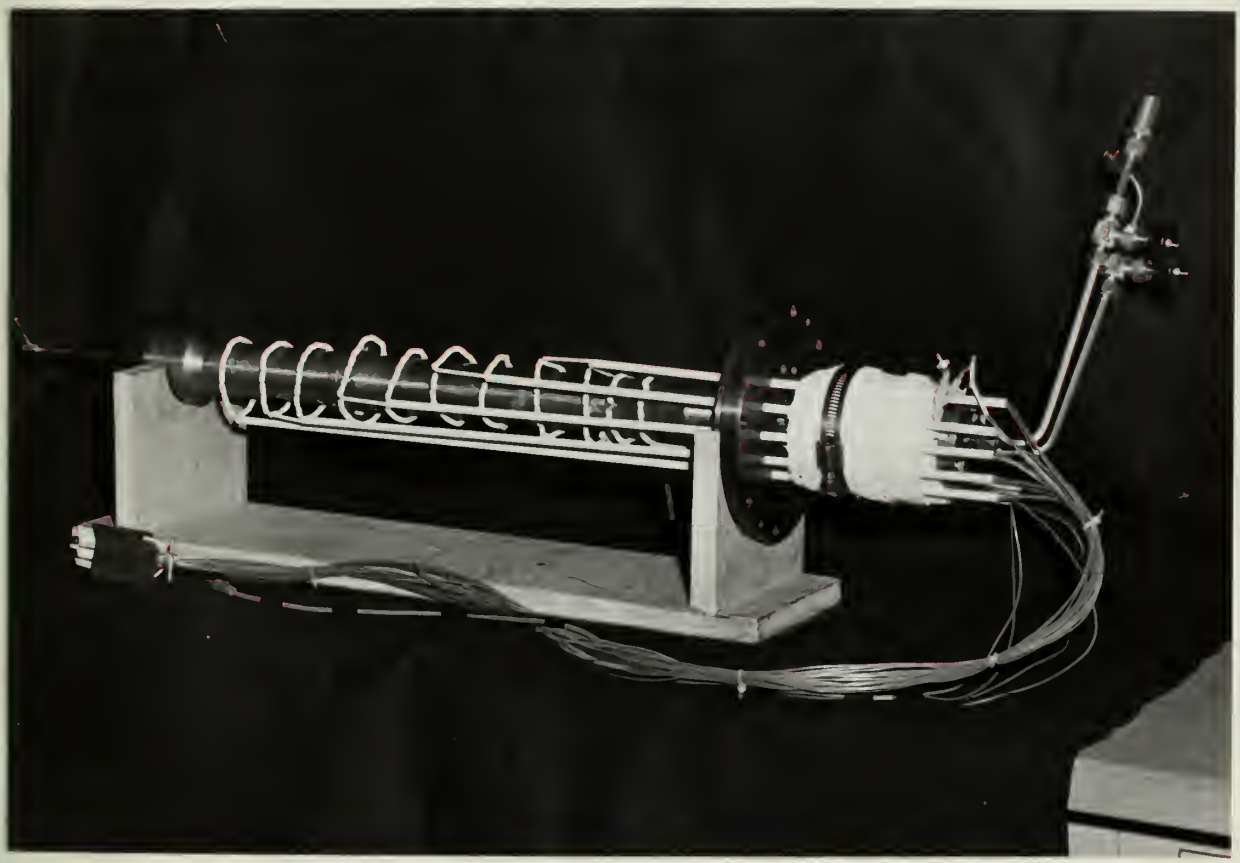

Fig. 2. Specimen bar assembly.

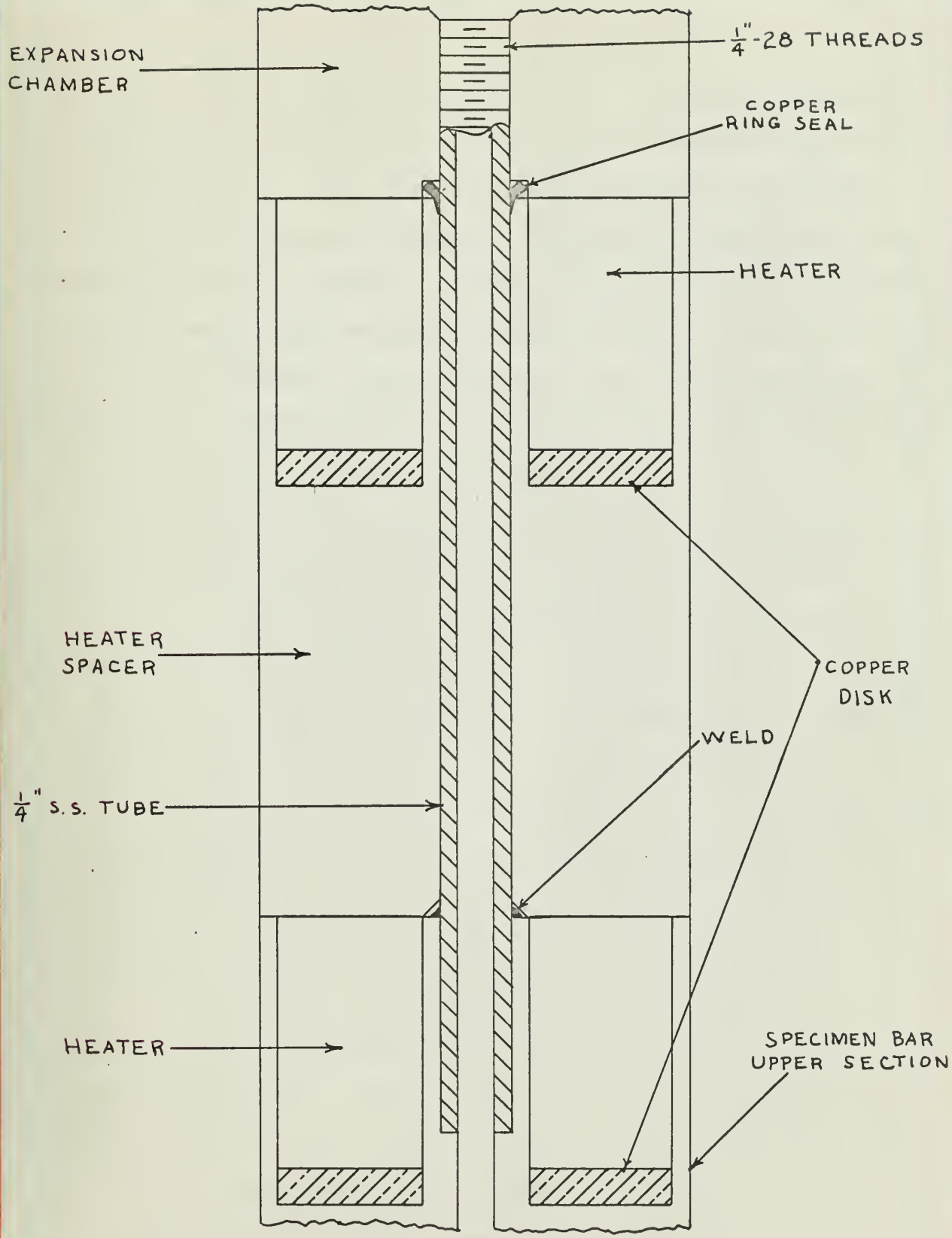

Fig. 3. Exploded cross-sectional view of upper portion of the specimen bar assembly, showing copper ring sealing arrangement.

heaters have 24 of these wire coils each 3/4 inch long, mounted vertically and evenly spaced on two concentric circles. This arrangement provides for an even distribution of heat across the specimen bar cross section. The melting chamber and drain tube heaters are of similiar construction but have 12 coils each 5 inches long which are evenly spaced on the heater circumference. The melting chamber heater is 3 inches in diameter while the drain tube heater has a 1 1/2-inch diameter. By using heaters of this type it was possible to manufacture them to fit the apparatus exactly, and they can be readily produced in the laboratory if replacement is necessary.

The temperatures in the specimen bar are measured by eleven chromel-alumel type thermocouples which are silver soldered into the bar at positions S 1 to S 11 in Fig. 1. A good quality wire manufactured by Leeds and Northrup Co. is specified to provide accuracies of $\pm 0.2^{\circ}C$. The thermocouples are calibrated using a platinum resistance thermometer in a well regulated high temperature oven and the temperatures measured are well within the limits of the wire itself. The thermocouple junctions at positions S 4 and S 5 are soldered into 1/64-inch indentations on the outer surface of the specimen chamber walls. This means that these two junctions are within 1/64 inch of the liquid metal sample, truly indicating the temperature therein. The thermocouples S 10 and S 11 measure the temperature at two points in the heater spacer. If these thermocouples have equal readings, then there is no upward heat loss from the lower specimen bar heater. The heat input to the specimen bar can then be found from the electric power input to the lower heater. All thermocouple leads are passed completely around the specimen bar to reduce conduction errors and then led out through Mullite* ceramic tubes which are supported by the top

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* Trade mark - McDanel Refractory Porcelain Co.

cover (12 in Fig. 1). A switching arrangement, which is described later, is designed so that all thermocouples are connected to a common reference junction and each can be selected when desired. Some difficulties were encountered when a few of the thermocouple leads broke off at the junction when subjected to high temperatures. These were replaced by merely resoldering new junctions in place of the broken ones. As yet, no method has been devised to prevent this breakage from occurring.

C. Guard Ring Assembly

The guard ring assembly (2 in Fig. 1) has an inside diameter of 5 1/2 inches, a wall thickness of 5/8 inch and is 20 inches long. It is machined in a single piece from a suitable tube of type 304 stainless steel. Heat is supplied to the guard ring by two Chromalox* type HBN 607 electric heater bands clamped in one inch grooves machined in its upper portion (13). Each of these grooves is lined with strips of copper to provide even heat distribution around the guard ring circumference. Cooling is provided at the base of the guard ring by two air ducts (14). The thermal conductivities of the liquid metals being tested are usually considerably greater than that of stainless steel, and consequently the thermal gradient in the specimen bar is reduced at the specimen chamber. Therefore, additional cooling and heating is provided for the guard ring in this same area (15) so that this reduced gradient can be matched. This heater is specially made by Chromalox for this purpose. Eleven chromel-alumel thermocouples are located in positions G 1 - G 11, which are exactly opposite those in the specimen bar at S1 - S11.

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* Trade mark - Edwin L. Wiegand Co.

If the temperatures of each pair of thermocouples (e.g., S 1 - G 1, S 2 - G 2 etc.) are equal there exists no heat flow between the specimen bar and the guard ring. The thermocouple leads are passed completely around the guard ring and then through Mullite protection tubes to the top of the furnace from whence they are connected to the switching circuit. Figure 4 is a photograph of the guard ring assembly showing heaters and thermocouples in place.

D. Furnace Core

Two 12 inch alundum cylinders (Norton type 50539) are cemented together coaxially to form a furnace core 10 inches in diameter, 24 inches long and having 1/2-inch thick walls, (16 in Fig. 1). The core is cast with double threads of 6 turns per inch pitch on its outer surface, which accommodate the main furnace heater windings. These heating coils are of No. 14 Kanthal A-1 wire, and are non-inductively wound in 10 separate segments F 1 - F 10, each corresponding to a pair of thermocouples in the guard ring and specimen bar. Each heater segment is individually controlled to assist in maintaining the guard ring temperature gradient, so that the furnace core itself acts as a second guard ring. As previously mentioned, the core is mounted in a rectangular enclosure of 1-inch Marinite whose outside dimensions are 14 inches by 14 inches by 24 inches. The space between the outer enclosure and the furnace core is filled with Norton type E-163 granular aluminum oxide insulation. Figure 5 shows the furnace core with heater windings in place. The heater leads are shown passing through one of the outer furnace walls.

E. Power Supplies

Two power supplies were constructed to provide power for the specimen heaters and the furnace windings. The main furnace power supply is pictured

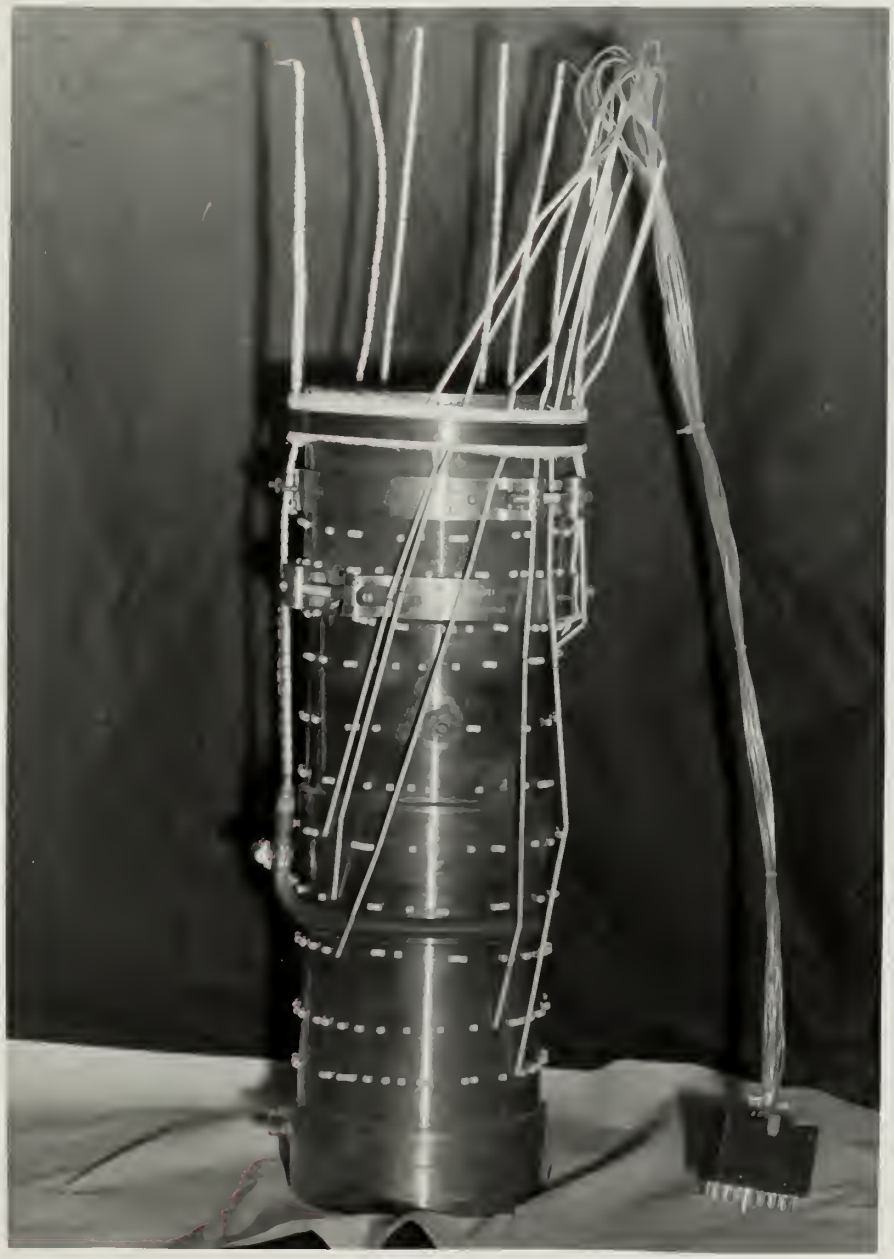

Fig. 4 Guard ring assembly.

in Fig. 6, and its circuit diagram is shown on Fig. 7. The ten variable transformers which control the ten sections of the furnace core heater are type Q-117-U Powerstats* each having a capacity of 1.2 KVA.** This power supply requires an input of two 115-volt 50-ampere circuits, and it was determined that this could be best provided by 2 phases of a 230-volt 3-phase line, each phase being 115 volts relative to neutral. The specimen bar heaters are powered by direct current to avoid creating any ac fields in the specimen bar. The dc power supply circuit is shown in Fig. 8, and consists of two bridge rectifier circuits with capacitor filters. It has a maximum power output of 60 watts for each heater.

The electrical power input to the specimen bar heaters is measured by Hewlett-Packard 3440A digital VTVM. A one ohm precision resistor (R 3, R 4) in series with each heater is used for current measurements. The selector SW 1 chooses which voltage is to be measured.

The guard ring, melting chamber and drain tube heaters are each powered by ordinary laboratory variable transformers, which give a sufficient degree of control and regulation for this purpose. It was found that because of the high heat capacity and low thermal conductivity of the stainless steel, small fluctuations in any of the heater supplies have little or no effect on the temperatures in the specimen bar and guard ring. A long term shift in line voltage (such as from day to night) does affect them somewhat, but all are changed by a similar amount.

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* Trade mark - Superior Electric Co.

** It was found later that this power supply is much more powerful than is actually needed to operate the furnace, for it has never been operated at greater than half of its full capacity. It would be suitable for supplying furnaces operating at much higher temperatures.

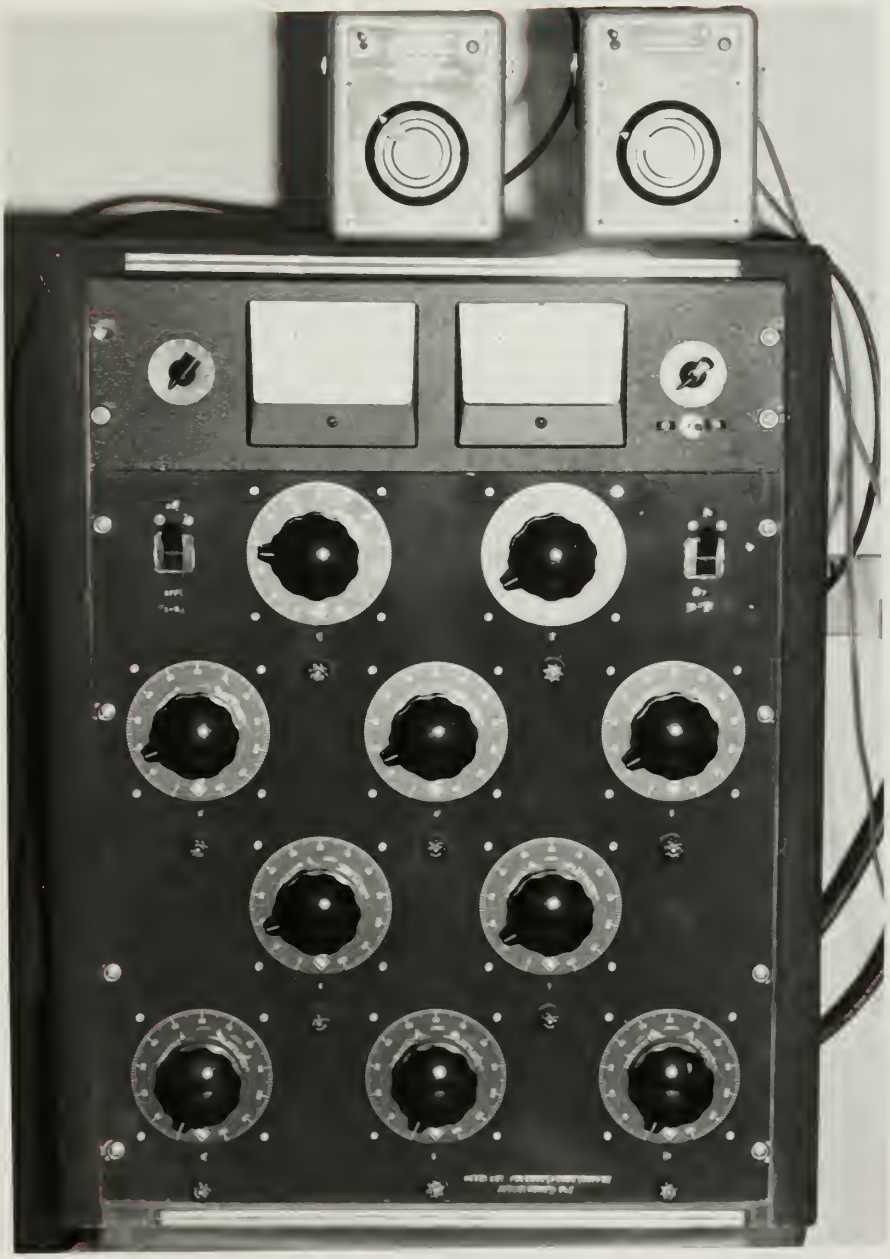

Fig. 6 Furnace power supply.

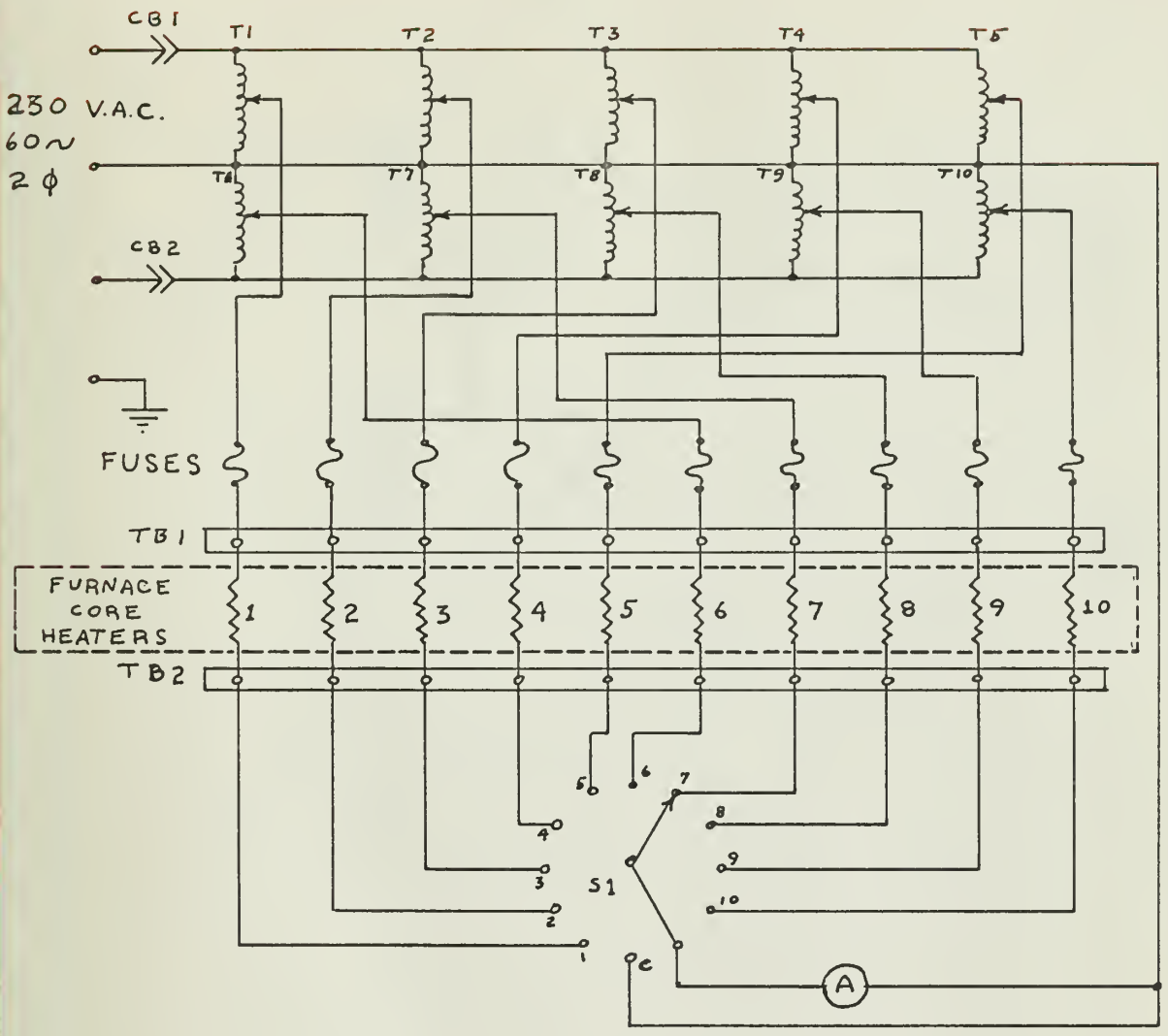

Fig. 7 AC power supply for furnace core heaters.

T1-T10	Powerstat type Q-117-U variable transformer
CB1, CB2	Circuit breaker, 60 amp.
Fuses	3AGC, 10 amp.
TB1, TB2	Terminal boards, high current barrier type
A	AC ammeter, 0-10 amp.
S1	Circuit opening switch, Mallory type 1400L. All terminals except that selected are shorted to a common terminal C.

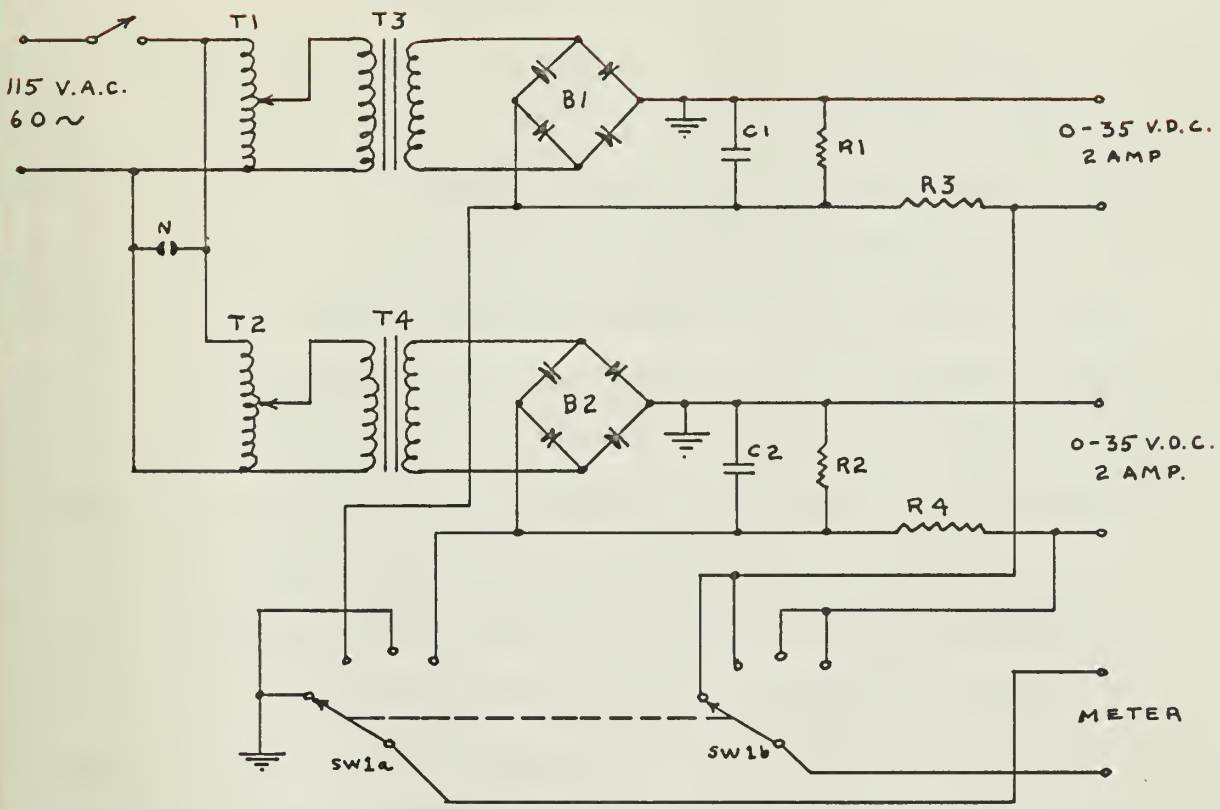

Fig. 8 DC Power Supply Circuit

T1, T2	Powerstat type 10B	R1, R2	2000 ohms, 1 watt
T3, T4	Stancor RT-204	R3	1 ohm, 0.05% 10 watt
B1, B2	Silicon bridge rectifier, RCA 1N1613 or TI 1N1616	R4	1 ohm, 1% 10 watt
		N	Neon bulb, 115 volt
C1, C2	500 µfd, 50 V.D.C.		

F. Thermocouple Monitoring Circuit

A circuit diagram of the thermocouple switching arrangement is shown in Fig. 9. Switch SS 2 has four positions marked specimen bar, guard ring, null and selector 3. A second selector switch SS 1 has eleven positions corresponding to the eleven thermocouples in the specimen bar and guard ring. If selector two is in either the specimen, guard or null position, then SS 1 will select which of the eleven thermocouples will be monitored. Thermocouple emf's are measured by a Hewlett-Packard 3440A digital voltmeter or a Leeds & Northrup type potentiometer, whichever is available. Use of the digital VTVM for this purpose allows all 22 thermocouple emf's to be read more rapidly, but the potentiometer has somewhat greater accuracy. The null position of SS 2 places the emf's of any selected thermocouple pair in series with opposite polarity so that a galvanometer in the circuit will be nulled when the emf's are equal. The fourth position of SS 2 allows for any thermocouple selected by SS 3 to be monitored, which would include the melting chamber and drain tube thermocouples. Except for null measurements, the thermocouple being monitored is connected to a common reference junction which is kept at $0^{\circ}C$ by a dewar of crushed ice and water. All other elements of the circuit are kept at room temperature to prevent errors in the generated emf's. The thermocouple control panel and dc power supply are pictured in Fig. 10.

G. Specimen Bar and Guard Ring Cooling

It has been previously mentioned that the guard ring and specimen bar are fitted with grooves or spaces for cooling air to be introduced. It was originally intended that water would be used as a coolant, but it was found that water cooled the specimen bar much too rapidly and very large gradients

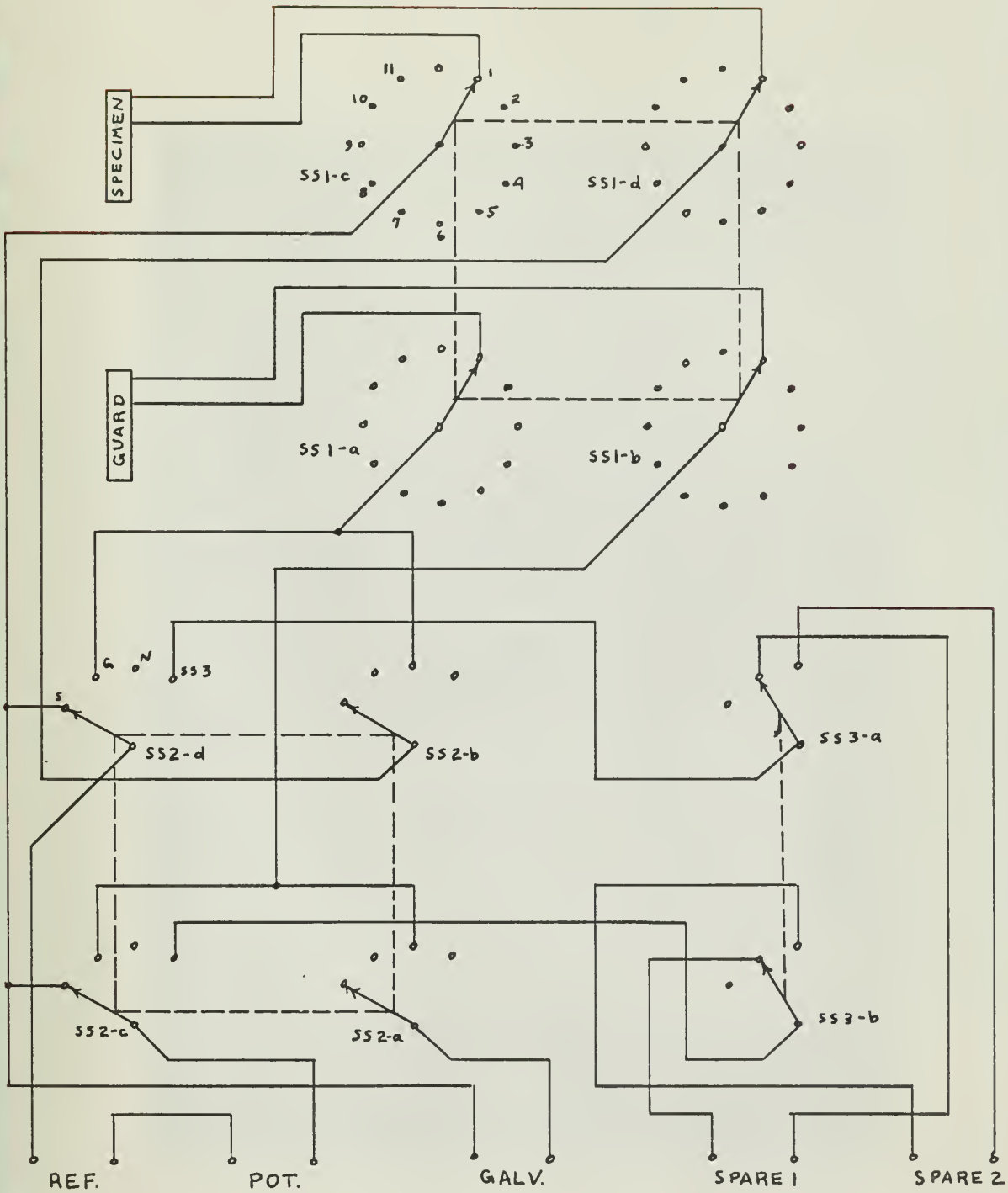

Fig. 9. Thermocouple Monitoring Circuit. Only two pairs of thermocouple leads are shown leading to SS 1. The others have been omitted for clarity.

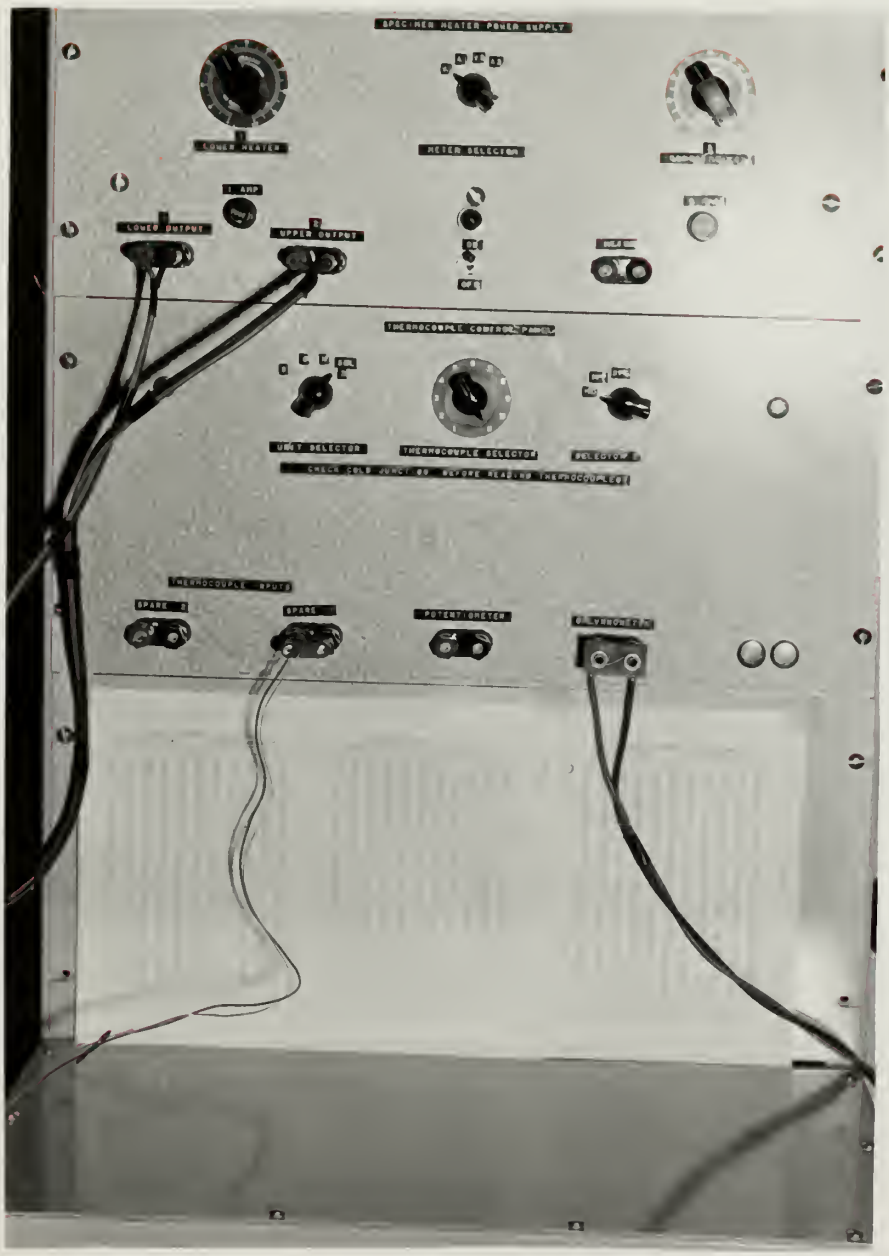

Fig. 10 Thermocouple control panel and dc power supply.

were produced. In addition, the water boiled immediately upon entering the cooling space, and this was not desirable. It was found that air at a pressure of 10 psi metered by Hoke "Milli-mite" needle valves provided the correct amount of cooling for both specimen bar and guard ring. Special micrometer handles on these valves indicate to the nearest 0.05 turn how far the valve is open. The 50-psi laboratory air line was used as a supply with a W. R. Brown Co. regulator and filter. In addition, a Hoke type 635-G4B 25-125 micron sintered bronze filter was installed in the line to keep tiny particles out of the cooling chambers.

H. Unit Assembly

The entire thermal conductivity apparatus is mounted on a 2 x 3 x 3 foot Dexion* frame with a shelf of 1-inch Marinite on which the assembly rests. The guard ring assembly is mounted first, held in place by four 1/4" - 20 bolts into the bottom cover. After connecting the 3/16-inch copper tubes which supply cooling air to the appropriate ducts, the furnace core (with windings already installed) is then placed around the guard ring. The stiff core heater leads which pass through one of the marinite outer walls hold the core in position. Alundum insulation is then packed between the outer furnace walls and the core, and the furnace is then complete. The specimen bar assembly, which has been previously tested under vacuum, is then lowered into the center of the guard ring and bolted in place by eight 1/4" - 20 allen cap screws in the top cover. The screws are coated with Never-Seez to prevent seizure when heated. A bottom cap (17 in Fig. 1) is then inserted on the drain tube and the drain tube heater is installed with a hose clamp.

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* Trade mark - Dexion Co.

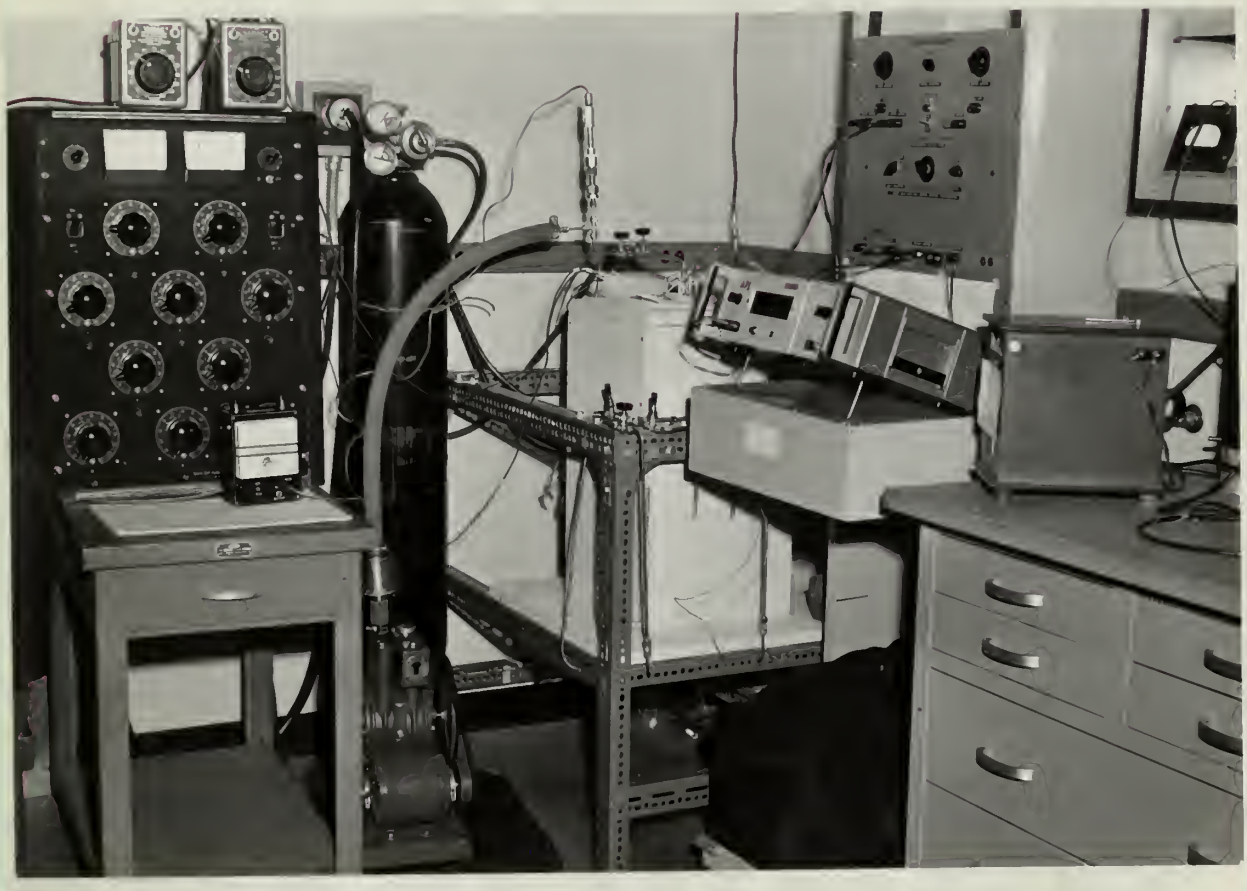

Fig. 11 Photo of entire experimental setup showing power supplies, furnace and thermocouple control panel.

The unit is shown completely assembled with power supplies in Fig. 11.

IV. EXPERIMENTAL PROCEDURE

A. Sample Preparation

In order to provide accurate results, a reasonable degree of care must be taken in the preparation of the metal samples to be tested. The volume of the specimen bar, including specimen chamber, drain and filler holes, is calculated from the dimensions to be about 66 cm^3. It is desired that sufficient metal be introduced so that the specimen bar will be filled to just below the expansion chamber, and this is done by preparing the proper amount of metal into one or more cylindrical ingots which will fit into the melting chamber. After determining the weight of metal to be used from its density, the pure metal sample is liquefied and passed through a fritted glass filter, which removes any foreign particles of a size greater than 50 microns. The metal is then cast in an ingot 1 5/8 inches in diameter and 2 inches long. It is then necessary to machine all exposed outside surfaces of the ingot to remove any oxides which may have formed as the metal solidified in its mold. The cleaned sample is then ready to introduce into the specimen chamber. For thermal conductivity measurements made on pure tin in this work, two ingots having a total weight of 500 grams were used.

B. Introducing the Metal Sample

Most metals will form oxides at a fairly rapid rate when liquefied in the presence of oxygen. When introducing the liquid metal sample to the specimen chamber, it is important that oxides or "slag" not be allowed to form on the surfaces, for the presence of these materials might significantly

affect the thermal conductivity of the sample and cause the metal to adhere to the interior surfaces of the specimen bar, especially the filling and drain tubes. To prevent the formation of oxides, purified samples are placed in the melting chamber as solids at room temperature. The melting chamber lid is then bolted in place, sealing off the entire specimen bar interior. The air in the specimen bar is then evacuated through one of the openings in the melting chamber lid, and the pressure is kept at 50 microns or less by continuous pumping. Using all heaters in the apparatus except the melting chamber heater, the entire specimen bar (except for the melting chamber) is raised above the melting point of the metal under test, so that when melted, the sample will fill the specimen bar down to the lower end of the drain tube. Energizing the melting chamber heater melts the sample; the sample then flows down into the heated, evacuated specimen bar interior. By monitoring thermocouples S 4 and S 5 as the sample is melted, it can be determined when the process is complete, for the thermal gradient between these thermocouples will drop sharply as the specimen chamber is filled. The apparatus is then ready to make thermal conductivity measurements. After the sample is introduced, the remainder of the specimen bar is filled with a dry, inert gas such as argon or nitrogen at a pressure of about 20 psi. This will allow the evacuation to be stopped and still prevent air from seeping in through any remaining system leaks. The system is kept pressurized with 20 psi of this gas throughout the measurements.

C. Thermal Conductivity Measurements

Perhaps the most important requirement for the precision of thermal conductivity measurements by the longitudinal heat flow method is the need for steady state conditions of heat flow and temperature in the specimen bar

and guard ring. It is also necessary to have equal temperatures at any given level or at any specified pair of thermocouples. These two conditions can only be set up by a careful adjustment of the various heaters, and cooling air valves and a great deal of patience. When the sample is in the specimen bar, the specimen heaters H 1 and H 2, the guard ring heaters, the specimen and guard cooling valves and the furnace core windings are set at an arbitrary position which might provide the proper gradients. The unit is then allowed to arrive at a thermal steady-state, which may take several hours. The 22 thermocouples S 1 - S 11 and G 1 - G 11 are read as often as necessary* to determine if a steady state is present. The readings of each pair of thermocouples are compared and appropriate adjustments made on heaters, cooling valves, etc., to match up the temperatures and gradients between specimen bar and guard ring. In addition, the power input to the lower specimen heater is recorded in each set of data. When equilibrium conditions are again reached, data are again recorded and the same process is repeated. This procedure is continued until the condition of no radial and upward heat loss at equilibrium exists. The thermal conductivity of the sample can then be computed using the power input, area and thermal gradient across the sample under these conditions. The mean temperature across the sample (i.e., the average of thermocouples S 4 and S 5) is the temperature corresponding to that particular thermal conductivity. For measurements at other temperatures, all heaters and cooling are adjusted a similar amount and the entire procedure is begun again. In this way, it is possible to make thermal conductivity measurements at many different temperatures as long as the two

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* The frequency of readings depends how close the unit is to the desired equilibrium condition.

necessary conditions are fulfilled in each case.

After having completed the measurements on a particular sample, the temperature of the entire specimen bar including the drain tube, is then raised above the melting point of the metal. The cap on the bottom of the drain tube is then removed, and the metal is allowed to flow out. The pressure of the inert gas in the melting chamber forces the liquid metal to drain out rapidly and completely. It is then advisable, after allowing the entire apparatus to cool to room temperature, to disassemble the specimen bar at the specimen chamber and check to see if any particles of the sample remain. The specimen bar is then cleaned and reassembled for the next run.

D. Experimental Results

The first test runs of the apparatus were made with pure* tin as a sample. This metal was selected because several investigators have made thermal conductivity measurements on this metal and their results are used as a check on the operation of the apparatus. The results obtained are shown in Fig. 12 and table I along with the results found in the literature. The tin sample in the second run had been filtered, whereas that used in the first had not, which would account for the difference in the results. A sample set of equilibrium data from which computations can be made is shown in the table below.

Thermocouple Emf's in millivolts

T/C#	1	2	3	4	5	6	7	8	9	10	11
S	10.35	10.92	11.47	11.96	12.31	12.86	13.32	13.79	14.50	14.80	14.80
G	10.37	10.91	11.44	11.95	12.35	12.88	13.32	13.84	14.51	14.49	14.23

* The actual purity of the samples used was unknown, but it was given by the source as better than 99%.

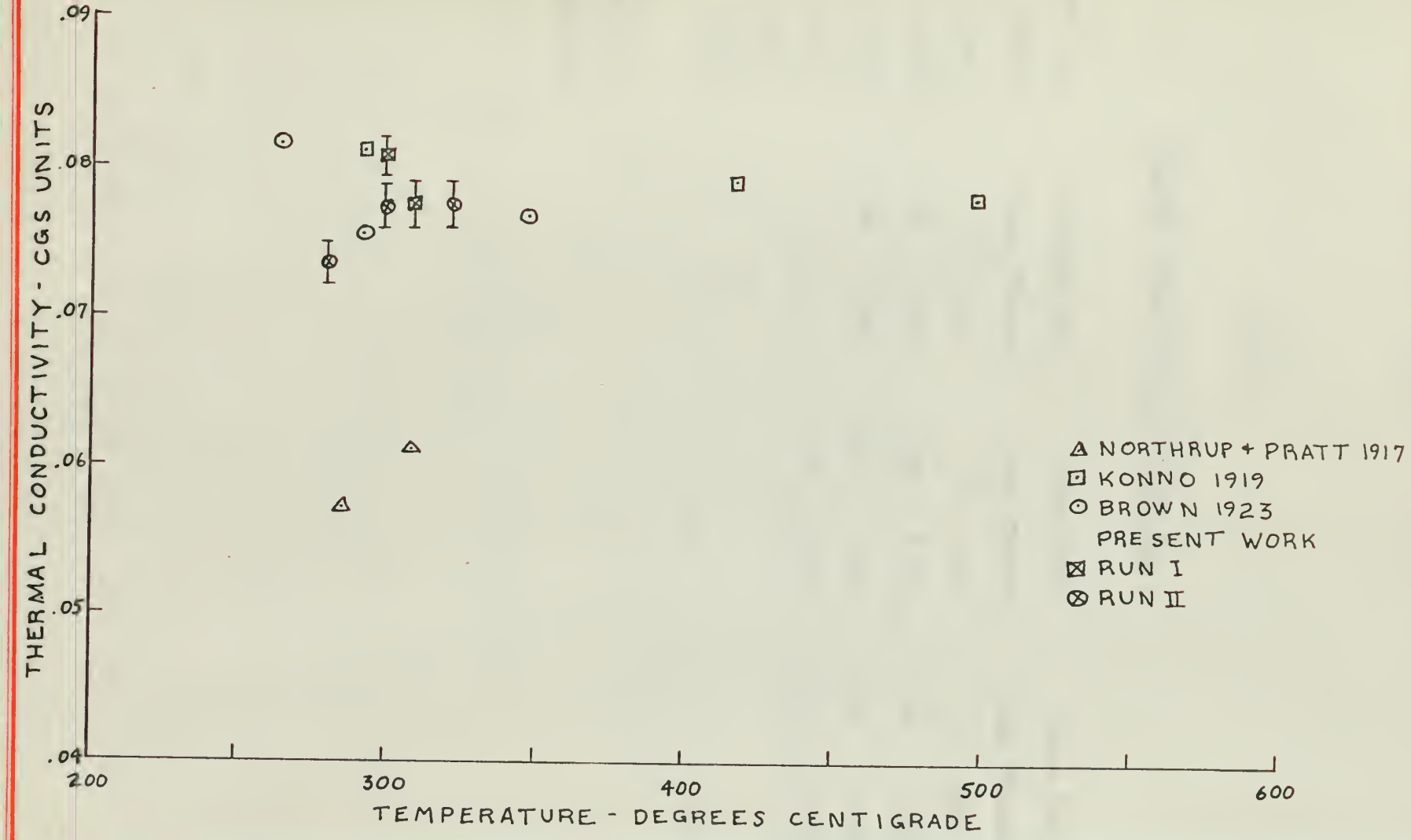

FIG. 12. THERMAL CONDUCTIVITY OF LIQUID TIN

Heater power input V = 14.9 volts, I = .653 amp.

Average temperature of sample = 298°C

Average outside diameter of specimen bar: 1.6258 inches*

Average inside diameter of specimen chamber: 1.5668 inches

Recalling Eq. (1),

$$Q = KA \frac{dT}{dx} .$$

From the specimen bar dimensions:

Cross sectional area of specimen bar (A_s) = 13.3944 cm^2

Cross sectional area of specimen chamber (A_T) = 12.4395 cm^2

Cross sectional area of specimen chamber walls (A_w) = 0.9549 cm^2

Electrical power input (Q_i) = 9.7297 watts = 2.3244 cal./sec.

First, the amount of heat conducted through the specimen chamber walls (Q_w) is found. The average temperature gradient in the steel is found to be 3.2152 degrees per cm. Hence

$$K_s = \frac{Q_i}{A_s \left(\frac{dT}{dx}\right)_s} = \frac{2.3244}{(13.3944)(3.2152)} = 0.0540 \text{ cgs units}$$

where K_s is the average thermal conductivity of the steel in the specimen bar at the sample temperature. From the sample data, the temperature difference between S4 and S5 is 35 millivolts or 8.4 degrees, and the distance between S4 and S5 is 1.5008 inches or 3.8120 cm and the thermal gradient across the specimen $\left(\frac{dT}{dx}\right)_T$ is $\frac{8.4}{3.8120}$ or 2.2036 degrees per cm. Therefore

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* Measured at room temperature, corrected for thermal expansion to 300°C.

$$Q_w = K_s A_w \left(\frac{dT}{dx}\right)_T = (.0540)(.9549)(2.2036) = 0.1137 \text{ cal./sec.}$$

The total heat conducted through the sample is

$$Q_T = Q_i - Q_w = 2.2107 \text{ cal./sec.}$$

The thermal conductivity of the liquid tin is then

$$K_T = \frac{Q_T}{A_T\left(\frac{dT}{dx}\right)_T} = \frac{2.2107}{(12.4395)(2.2036)} = 0.0806 \text{ cgs units}$$

This is the thermal conductivity at 298°C, the average temperature of the specimen chamber.

V. DISCUSSION OF RESULTS

A. Error Analysis

In any scientific measurement, it is important to determine the accuracy of the results. In the thermal conductivity measurements described here, errors occur in three quantities: The heat input to the specimen bar, the area of the specimen bar and the thermal gradient. It is convenient to consider each of these individually.

The electrical power input to the sample is determined by measuring the input voltage and current to the lower specimen bar heater. The voltage is measured directly and the current is found by determining the voltage drop across a one-ohm precision resistor. The accuracy of the voltmeter used is

given by the manufacturer as ±0.05% ± 1 digit and the precision resistor tolerance is also ±0.05%. This means that the measurements of input voltage can deviate as much as 0.05% and the current as much as 0.1% due to the measuring devices. However, since the dc heater power supply is not highly regulated the voltage fluctuates as much as ±0.05 volt and the current deviates by ±0.005 amp. The error in input voltage is then $\frac{0.05}{15} \approx 0.3\%$ and in the current $\frac{0.005}{0.5} = 1\%$. The meter error can therefore be neglected since it is small compared to the above, and the total error in power input is $\sqrt{1^2 + (0.3)^2} = 1\%$. Inspection of the sample data given previously shows that the emf's of some thermocouple pairs are not perfectly matched, indicating a small heat flow between specimen bar and guard ring or vice versa. A difference of 0.04 millivolt corresponds to one degree of temperature difference in this temperature range. Because of the low conductivity of the air in the annular space between the specimen bar and guard ring, and the small temperature difference of one degree or less the heat lost or gained is very small and is estimated to be less than 1% of the total heat input. The total error in the amount of heat conducted through the sample is therefore about 2%.

In the determination of the thermal gradient $(\frac{dT}{dx})$, errors occur in the measurement of temperature and the distance between thermocouples. Temperature errors come from the device used to measure the thermocouple emf's and the thermocouple wire itself. The Hewlett-Packard 3440A digital VTVM used to measure emf's is accurate to ±0.1% of reading ± 1 digit on the 100 millivolt scale. Since all emf's are less than 20 mv., the maximum deviation is 0.001 x 20 + 0.01 = 0.03 mv, which corresponds to an error of $\frac{0.03}{20} = 0.15\%$. The

thermocouple wire is specified to be accurate to $\pm 0.2^{\circ}C$, and for a temperature of $300^{\circ}C$ this corresponds to 0.05% error. The quantity used in the calculations is a temperature difference therefore this error is doubled, for the temperatures at S4 and S5 can both be in error by this amount. An additional error of 0.1% will be added by the deviation of the thermocouple reference junction from $0^{\circ}C$. The mounting holes for the thermocouples in the specimen bar were located and drilled using a precision indexing machine and the maximum deviation in the distance between holes is .002 inch. For a distance between thermocouples of 1.5 inches, the error is $\frac{.004}{1.5} = 0.3\%$. The total error in the measurement of the thermal gradient is therefore less than 0.5%.

The diameters of the specimen bar and chamber are measured with a precision micrometer at several positions and averaged together. Since the micrometer has a maximum deviation of 0.001 inch, and measured diameters are squared in the calculation of cross sectional area, the deviation must be multiplied by 2. The resulting error in cross sectional area is $\frac{.002}{12} = 0.2\%$.

Since the quantities Q, A and $(\frac{dT}{dx})$ are multiplied together, the total measurement error is the square root of the sum of the squares of the errors in each measured quantity. Hence

$$\text{Total error} = \sqrt{(2)^2 = (0.5)^2 + (0.2)^2} = 2.05$$

or slightly over 2%. It is evident that the greatest source of error is in the determination of heat input to the sample due to fluctuations in heater input power and radial losses (or gains) between specimen bar and guard ring.

Other possible sources of error are found in the conduction losses along

the thermocouple wires, heat transfer by convection between specimen bar and guard ring and non-equilibrium heat flow conditions. The first of these has been reduced to a very small quantity by wrapping the thermocouple wires completely around the specimen bar or guard ring before passing them out of the apparatus. Any remaining conduction losses are considered negligible. Since the specimen bar and guard ring are both kept warmer at the top, it is unlikely that any convection currents can exist between them, and this error source is ruled out. When a set of equilibrium data was taken, after allowing many hours for equilibrium to be achieved, the thermocouple emf's were always constant within ±0.01 millivolt over the period of the measurements (about 5 minutes). Since this fluctuation is less than the inherent error of the measuring instrument, it can be neglected.

B. Improvements

Two major improvements, which should improve the accuracy and ease of disassembly of the apparatus, are apparent at this writing. To eliminate fluctuations in the output of the dc heater supply, a suitable regulating device should be employed. This device might utilize a sampling resistor and a feedback circuit so that the output current, or voltage can be kept constant. Circuits of this type can be found in the literature[15] and can be constructed at moderate cost. Use of this device would certainly reduce small changes in sample heat input and significantly reduce the error in this quantity. A second improvement in the construction of the thermocouple protection tubes would facilitate the disassembly of the specimen bar for cleaning between runs. The change would provide a break in the thermocouple leads at the end of each ceramic protection tube, and a plug-in system for

attaching the wires together. In the present system, the wires from each junction pass unbroken from the junction to the control circuit, and disassembly necessitates drawing the four foot extension wires out through the protection tubes. A plug-in arrangement shown in Fig. 13 can be made using Amphenol "Relia-tac" contacts crimped on to the thermocouple leads and mounted in the protection tubes.

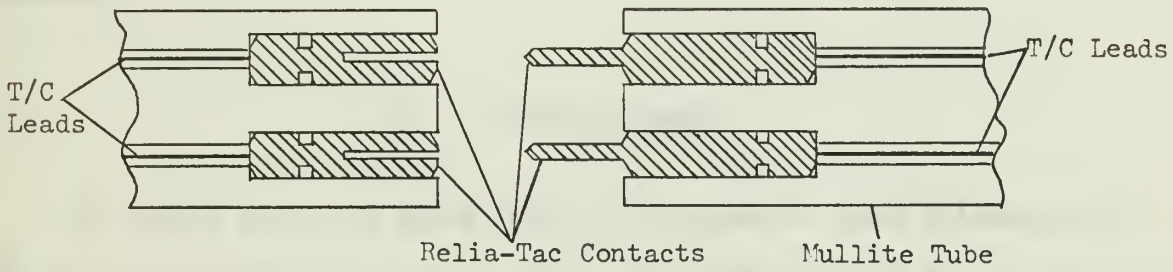

Fig. 13. Plug-in arrangement for thermocouple wires.

The most suitable ceramic tubes for this purpose would be McDanel type 4T36414 which are 1/4 inch O.D. and have four 3/64 inch bores. The holes must be counterbored to a depth of 1/2 inch with an 0.050 inch drill to accommodate the connectors as shown in Fig. 13.

The thermal conductivity apparatus described in this paper is currently capable of measurements on any liquid metal at temperatures up to 400°C., provided the metal does not corrode type 304 stainless steel. The operation could be expanded to include all metals by fabricating a specimen bar of a non-corroding material for the metal to be tested. The temperature limitation can be increased by using a higher temperature vacuum seal than the asbestos gaskets presently in use. The furnace power supply has sufficient power to provide temperatures far in excess of the present limit.

VI. CONCLUSION

It is hoped that the work described in this paper may provide a reliable method of measuring the thermal conductivity of any liquid metal or alloy at any temperature below the maximum limit. It is felt that of the methods available for making this type of measurement the one chosen is the most accurate.

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